

Theory of ultrafast nonequilibrium dynamics in d -wave superconductors

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We use density-matrix theory to calculate the ultrafast dynamics of unconventional superconductors from a microscopic viewpoint. We calculate the time evolution of the optical conductivity as well as pump-probe spectra for a d -wave order parameter. Three regimes can be distinguished in the spectra. The Drude response at low photon energies is the only one of those which has been measured experimentally so far. At higher energies, we predict two more regimes: the pair-breaking peak, which is reduced as Cooper-pairs are broken up by the exciting pulse; and a suppression above the pair-breaking peak due to nonequilibrium quasiparticles. Furthermore, we consider the influence of the electron-phonon coupling, and derive rate equations which have been widely used so far.

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Introduction – In recent years, numerous studies of the nonequilibrium dynamics of carriers in superconductors have been performed using femtosecond time-resolved spectroscopy [1, 2, 3, 4, 5, 6, 7]. In a typical experiment, the sample is excited with an intense fs laser pulse (pump pulse), and after a delay time Δt , spectra are measured using a second, less intense, laser pulse (probe pulse). As the nature of the interactions between quasiparticles in the high- T_c cuprates is still under debate [8], it is interesting to directly observe the characteristic dynamics of condensate depletion and Cooper-pair recombination. This can be done with real-time optical techniques. In the high- T_c superconductor $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ (BSCCO), for example, relaxation times of about 50 ps have been measured [4]; the observed decay is two-component (biexponential).

Theoretical attempts to model these experiments have, on the one hand, used quasi-equilibrium models (so-called μ^* , T^* models, [9]) to describe the state excited by the pump pulse. On the other hand, rate equation approaches based on the phenomenological Rothwarf-Taylor model [10] have been used [1, 4, 7] to describe the recovery dynamics of the superconducting state. It is assumed that the dynamics are governed by the creation of high-energy phonons due to Cooper-pair recombination and subsequent phonon decay. So far, there has been no attempt to describe the excitation *and* relaxation dynamics on equal footing. As well, no microscopic description of the related time dynamics is available.

In this Letter, we present a theory which can describe the femtosecond excitation and relaxation processes from a microscopic viewpoint. In particular, we consider high- T_c cuprates, using a realistic band structure and considering coupling to two important phonon modes (breathing and buckling modes, which are strongly coupled to the superconducting CuO_2 planes [11]). We employ the approach of density-matrix theory, which has been used to

some extent to describe ultrafast dynamics in semiconductors (see e.g. Ref. [12, 13, 14]).

Theory – We start from a Hamiltonian $H = H_{\text{sc}} + H_{\text{field}} + H_{\text{phon}}$, where H_{sc} describes the superconducting state, H_{field} gives the interaction with the classical electromagnetic field, and H_{phon} models the bare phonons and their interaction with the electrons. Explicitly we write

$$H_{\text{sc}} = \sum_{\mathbf{k}s} (\epsilon_{\mathbf{k}} - \mu) c_{\mathbf{k}s}^+ c_{\mathbf{k}s} + \sum_{\mathbf{k}} \left(\Delta_{\mathbf{k}} c_{\mathbf{k}\uparrow}^+ c_{-\mathbf{k}\downarrow}^+ + \text{h.c.} \right), \quad (1)$$

$$H_{\text{field}} = -\frac{e\hbar}{m} \sum_{\mathbf{k}\mathbf{q}s} (\mathbf{k} \cdot \mathbf{A}_{\mathbf{q}}) c_{\mathbf{k}+\frac{\mathbf{q}}{2}s}^+ c_{\mathbf{k}-\frac{\mathbf{q}}{2}s} + \frac{e^2}{2m} \sum_{\mathbf{k}s} (\mathbf{A}_{\mathbf{q}-\mathbf{k}} \cdot \mathbf{A}_{\mathbf{q}}) c_{\mathbf{k}s}^+ c_{\mathbf{k}s}, \quad \text{and} \quad (2)$$

$$H_{\text{phon}} = \sum_{\mathbf{q}j} \hbar\omega_{\mathbf{q}j} \left(b_{\mathbf{q}j}^+ b_{\mathbf{q}j} + \frac{1}{2} \right) + \sum_{\mathbf{p}\mathbf{j}\mathbf{k}s} \left(g_{\mathbf{p}\mathbf{k}j s} (b_{-\mathbf{p}j}^+ + b_{\mathbf{p}j}) c_{\mathbf{k}+\mathbf{p},s}^+ c_{\mathbf{k}s} + \text{c.c.} \right). \quad (3)$$

In Eq. (1), $\epsilon_{\mathbf{k}}$ is a tight-binding band structure as measured by Kordyuk *et al.* [15], μ is the chemical potential, and $\Delta_{\mathbf{k}} = \Delta_0 (\cos k_x - \cos k_y)/2$ denotes a d -wave order parameter with $\Delta_0 = 30$ meV. c^+ and c are the electronic creation and annihilation operators, respectively. The j index counts the different phonon modes. In Eq. (2), $\mathbf{A}_{\mathbf{q}}$ denotes the Fourier component of the vector potential the superconductor interacts with. It includes both the pump and probe fields; as the interaction with the pump field is nonlinear, the quadratic terms in \mathbf{A} are needed. H_{phon} in Eq. (3) includes the bare phonons, having the dispersion $\omega_{\mathbf{q}j}$, and the electron-phonon interaction, described by the coupling matrix elements $g_{\mathbf{p}\mathbf{k}j s}$. b^+ and b are the creation and annihilation operators for the phonons. We consider the important breathing and buckling phonon modes [16]. These two modes are thought to be most

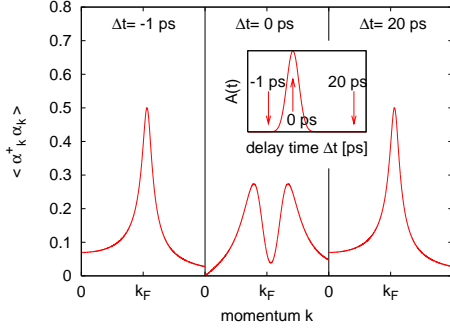


FIG. 1: Plot of the excitation process. Starting with an equilibrium quasiparticle distribution $\langle \alpha_{\mathbf{k}}^+ \alpha_{\mathbf{k}} \rangle$ before the pump pulse (left panel), a nonequilibrium distribution is excited (middle panel), which then relaxes back into equilibrium (right panel). The inset shows the exciting pulse, a 50 fs Gaussian. The \mathbf{k} -vectors lie in the 2D CuO_2 plane; the plot shows a cut along the antinodal direction, from $(0, \pi)$ to (π, π) , crossing the Fermi level at k_F . The temperature is 4 K.

strongly coupled to the superconducting state, and thus the most relevant for scattering processes which can lead to relaxation of excited quasiparticles.

We first perform a Bogoliubov transformation $\alpha_{\mathbf{k}}^+ = u_{\mathbf{k}} c_{\mathbf{k}\uparrow}^+ - v_{\mathbf{k}} c_{-\mathbf{k}\downarrow}$, $\beta_{\mathbf{k}}^+ = u_{\mathbf{k}} c_{-\mathbf{k}\downarrow} + v_{\mathbf{k}} c_{\mathbf{k}\uparrow}^+$. Within the Heisenberg picture we calculate equations of motion for the Bogoliubov quasiparticle densities $\langle \alpha_{\mathbf{k}_1}^+ \alpha_{\mathbf{k}_2} \rangle(t)$, $\langle \beta_{\mathbf{k}_1}^+ \beta_{\mathbf{k}_2} \rangle(t)$, which correspond to the excited states of a superconductor, and the anomalous expectation values $\langle \alpha_{\mathbf{k}_1}^+ \beta_{\mathbf{k}_2}^+ \rangle$, which correspond to the condensate of Cooper-pairs. The current density is then given by

$$\begin{aligned} \langle \mathbf{j} \rangle(\mathbf{q}, t) = & \frac{e\hbar}{m} \sum_{\mathbf{k}} (2\mathbf{k} - \mathbf{q}) \\ & \times \left[(u_{\mathbf{k}+\mathbf{q}} u_{\mathbf{k}} + v_{\mathbf{k}+\mathbf{q}} v_{\mathbf{k}}) (\langle \alpha_{\mathbf{k}+\mathbf{q}}^+ \alpha_{\mathbf{k}} \rangle - \langle \beta_{\mathbf{k}}^+ \beta_{\mathbf{k}+\mathbf{q}} \rangle) \right. \\ & + (u_{\mathbf{k}+\mathbf{q}} v_{\mathbf{k}} - v_{\mathbf{k}+\mathbf{q}} u_{\mathbf{k}}) (\langle \alpha_{\mathbf{k}+\mathbf{q}}^+ \beta_{\mathbf{k}}^+ \rangle - \langle \alpha_{\mathbf{k}} \beta_{\mathbf{k}+\mathbf{q}} \rangle) \left. \right] \\ & - \frac{e^2}{2m} \sum_{\mathbf{k}} \mathbf{A}_{\mathbf{q}-\mathbf{k}} \left(2v_{\mathbf{k}}^2 - \frac{\epsilon_{\mathbf{k}}}{E_{\mathbf{k}}} (\langle \alpha_{\mathbf{k}}^+ \alpha_{\mathbf{k}} \rangle + \langle \beta_{\mathbf{k}}^+ \beta_{\mathbf{k}} \rangle) \right). \end{aligned} \quad (4)$$

The first and last terms include Bogoliubov quasiparticle densities, thus describing the contribution of the normal part in a two-fluid-model. The second term, including anomalous expectation values, describes the condensate response. Both Bogoliubov quasiparticle densities and anomalous expectation values can be calculated for a given delay time Δt . As the probe field $E_{\text{probe}} = -i\omega A_{\text{probe}}$ is known, the optical conductivity σ can be calculated via $\langle j \rangle(\mathbf{q}, \omega) = -i\omega \sigma(\mathbf{q}, \omega) A_{\text{probe}}(\mathbf{q}, \omega)$. Only the \mathbf{q} -independent conductivity $\sigma(\mathbf{q} \rightarrow 0, \omega)$ will be considered.

Equations of motion – In order to calculate σ , the equations of motion for the Bogoliubov quasiparticle distributions and anomalous expectation values have to be solved. They both couple to phonon-assisted quantities

e.g. $\langle \alpha_{\mathbf{k}_1+\mathbf{q}}^+ \alpha_{\mathbf{k}_2} (b_{-\mathbf{q}j}^+ + b_{\mathbf{q}j}) \rangle$. We now use second-order cluster expansion [12, 17] and calculate equations of motion for the phonon-assisted quantities, which couple to 4-point quantities such as $\langle \alpha_{\mathbf{k}+\mathbf{q}}^+ \alpha_{\mathbf{k}}^+ \alpha_{\mathbf{k}+\mathbf{q}} \alpha_{\mathbf{k}} \rangle$. At this point, the hierarchy is broken down by factorizing the 4-point quantities. The phonons are assumed to remain equilibrated (bath approximation, $\langle b_{\mathbf{q}j}^+ b_{\mathbf{q}j} \rangle \rightarrow n_{\mathbf{q}j}$ with the Bose distribution $n_{\mathbf{q}j}$) while the quasiparticles are excited and relax. The equations for the phonon-assisted quantities can then be solved, giving rise to a system of integro-differential equations. For example, the equation for the Bogoliubov quasiparticle occupation $\langle \alpha_{\mathbf{k}}^+ \alpha_{\mathbf{k}} \rangle$ reads:

$$\begin{aligned} \partial_t \langle \alpha_{\mathbf{k}}^+ \alpha_{\mathbf{k}} \rangle = & -\frac{ie}{m} \mathbf{k} \cdot \mathbf{A}_{\mathbf{q}} M_{\mathbf{kq}} (\langle \alpha_{\mathbf{k}} \beta_{\mathbf{k}} \rangle - \langle \alpha_{\mathbf{k}}^+ \beta_{\mathbf{k}}^+ \rangle) \\ & + \sum_{\mathbf{q}j} \int_0^\infty ds \frac{\pi |g_{\mathbf{q}j}|^2}{\hbar^2} \left[(1 + n_{\mathbf{q}j}) e^{i(\omega_{\mathbf{k}+\mathbf{q}} - \omega_{\mathbf{k}} + \omega_{\mathbf{q}j})s} \right. \\ & \times L_{\mathbf{kq}} u_{\mathbf{k}} u_{\mathbf{k}+\mathbf{q}} \langle \alpha_{\mathbf{k}}^+ \alpha_{\mathbf{k}} \rangle(t-s) (1 - \langle \alpha_{\mathbf{k}+\mathbf{q}}^+ \alpha_{\mathbf{k}+\mathbf{q}} \rangle(t-s)) \\ & - n_{\mathbf{q}j} e^{i(\omega_{\mathbf{k}+\mathbf{q}} - \omega_{\mathbf{k}} - \omega_{\mathbf{q}j})s} \\ & \times L_{\mathbf{kq}} u_{\mathbf{k}} u_{\mathbf{k}+\mathbf{q}} \langle \alpha_{\mathbf{k}+\mathbf{q}}^+ \alpha_{\mathbf{k}+\mathbf{q}} \rangle(t-s) (1 - \langle \alpha_{\mathbf{k}}^+ \alpha_{\mathbf{k}} \rangle(t-s)) \\ & + e^{i(\omega_{\mathbf{k}+\mathbf{q}} + \omega_{\mathbf{k}} - \omega_{\mathbf{q}j})s} \\ & \left. \times M_{\mathbf{kq}} u_{\mathbf{k}+\mathbf{q}} v_{\mathbf{k}} \langle \beta_{\mathbf{k}+\mathbf{q}}^+ \beta_{\mathbf{k}+\mathbf{q}} \rangle(t-s) \langle \alpha_{\mathbf{k}}^+ \alpha_{\mathbf{k}} \rangle(t-s) \right] \end{aligned} \quad (5)$$

with $L_{\mathbf{kq}} = u_{\mathbf{k}+\mathbf{q}} u_{\mathbf{k}} + v_{\mathbf{k}+\mathbf{q}} v_{\mathbf{k}}$, $M_{\mathbf{kq}} = u_{\mathbf{k}+\mathbf{q}} v_{\mathbf{k}} - v_{\mathbf{k}+\mathbf{q}} u_{\mathbf{k}}$ being the relevant matrix elements, and $\omega_{\mathbf{p}} = E_{\mathbf{p}}/\hbar$, where $E_{\mathbf{p}} = \sqrt{\epsilon_{\mathbf{p}}^2 + \Delta_{\mathbf{p}}^2}$ is the Bogoliubov quasiparticle dispersion. There are 4 equations for the 4 expectation values appearing in Eq. (4), all with a similar structure. The full system is published elsewhere [18]. On this level, the equations are similar to the ones obtained within the Keldysh formalism, with the difference that here the *nonequilibrium* distributions $\langle \alpha_{\mathbf{k}}^+ \alpha_{\mathbf{k}} \rangle(t-s)$ with their full time-dependences contribute.

By using the Markovian approximation [19], the integrals can be solved and one finds, for example,

$$\begin{aligned} \partial_t \langle \alpha_{\mathbf{k}}^+ \alpha_{\mathbf{k}} \rangle = & -\frac{ie}{m} \mathbf{k} \cdot \mathbf{A}_{\mathbf{q}} M_{\mathbf{kq}} (\langle \alpha_{\mathbf{k}} \beta_{\mathbf{k}} \rangle - \langle \alpha_{\mathbf{k}}^+ \beta_{\mathbf{k}}^+ \rangle) \\ & + \sum_{\mathbf{q}j} \frac{\pi |g_{\mathbf{q}j}|^2}{\hbar^2} \left(\Gamma_{\mathbf{kq}j}^{(1)} \langle \alpha_{\mathbf{k}}^+ \alpha_{\mathbf{k}} \rangle (1 - \langle \alpha_{\mathbf{k}+\mathbf{q}}^+ \alpha_{\mathbf{k}+\mathbf{q}} \rangle) \right. \\ & - \Gamma_{\mathbf{kq}j}^{(2)} \langle \alpha_{\mathbf{k}+\mathbf{q}}^+ \alpha_{\mathbf{k}+\mathbf{q}} \rangle (1 - \langle \alpha_{\mathbf{k}}^+ \alpha_{\mathbf{k}} \rangle) \\ & \left. - \Gamma_{\mathbf{kq}j}^{(3)} \langle \beta_{\mathbf{k}+\mathbf{q}}^+ \beta_{\mathbf{k}+\mathbf{q}} \rangle \langle \alpha_{\mathbf{k}}^+ \alpha_{\mathbf{k}} \rangle \right), \end{aligned} \quad (6)$$

with

$\Gamma_{\mathbf{kq}j}^{(1)} = (1 + n_{\mathbf{q}j}) u_{\mathbf{k}+\mathbf{q}} u_{\mathbf{k}} L_{\mathbf{kq}} \delta(\omega_{\mathbf{k}+\mathbf{q}} - \omega_{\mathbf{k}} + \omega_{\mathbf{q}j})$,
 $\Gamma_{\mathbf{kq}j}^{(2)} = n_{\mathbf{q}j} u_{\mathbf{k}+\mathbf{q}} u_{\mathbf{k}} L_{\mathbf{kq}} \delta(\omega_{\mathbf{k}+\mathbf{q}} - \omega_{\mathbf{k}} - \omega_{\mathbf{q}j})$,
 $\Gamma_{\mathbf{kq}j}^{(3)} = u_{\mathbf{k}+\mathbf{q}} v_{\mathbf{k}} M_{\mathbf{kq}} \delta(\omega_{\mathbf{k}+\mathbf{q}} - \omega_{\mathbf{k}} + \omega_{\mathbf{q}j})$. This is a Boltzmann-type equation describing both in- and out-scattering with phonons ($\Gamma_{\mathbf{kq}j}^{(1)}$, $\Gamma_{\mathbf{kq}j}^{(2)}$) and Cooper-pair recombination ($\Gamma_{\mathbf{kq}j}^{(3)}$) processes. Finally, numerical solution yields the Bogoliubov quasiparticle distributions

and anomalous expectation values, and thus the optical conductivity.

Results – Exciting the initial Bogoliubov quasiparticle distribution $\langle \alpha_{\mathbf{k}}^\dagger \alpha_{\mathbf{k}} \rangle = f(E_{\mathbf{k}}) = (1 + \exp(E_{\mathbf{k}}/k_B T))^{-1}$ with a fs pump pulse, a nonequilibrium distribution is created, as shown in Fig. 1. The biggest changes are around the Fermi energy, and the distribution is clearly non-thermal – quasiparticle weight is rearranged and consequently the condensate is also in a non-thermal state. Because of scattering with phonons, this nonequilibrium distribution can subsequently relax back into an equilibrated one.

The probe conductivity after the pump and pump-probe spectra are obtained using the calculated Bogoliubov quasiparticle distributions as shown in Fig. 2. We can identify three regimes: the low-energy part (I) shows the Drude response, i.e. the response of the normal part in a two-fluid-model. The low-frequency power laws for d -wave superconductors are still obeyed after excitation. As Cooper-pairs are broken up by the pump pulse, thus generating Bogoliubov quasiparticles, the Drude response gets stronger. At higher energies $\approx 2\Delta_0$ one finds the pair-breaking peak (II). It gets shifted after pumping, as the superconducting state is depleted and Cooper-Pairs are broken up. Above the pair-breaking peak (region III), the absorption, $\alpha \sim \sigma_1/\omega$, is suppressed. In an absorption process, Cooper-pairs have to be broken up, and the generated quasiparticles have to have empty states above $2\Delta_0$ to be excited into. As a large number of quasiparticles are already excited due to the pump process, there are less states available than at equilibrium, which decreases the absorption. So far, only the Drude response part (I) has been measured experimentally [4]. In principle, however, the regimes II and III could be measured in THz pump-THz probe experiments.

The enhancement of the Drude response and the shift of the pair-breaking peak are also found in a T^* model, where the excited quasiparticle distribution is assumed to be an equilibrium distribution with an effective temperature T^* [9]. However, the suppression above $2\Delta_0$ is not found within a T^* model (see Fig. 3). It is a nonequilibrium effect – simply speaking, enhancing the temperature does not create enough Bogoliubov quasiparticles to fill a large number of states above $2\Delta_0$.

Apart from pump-probe spectra, one can also look at the time evolution of the optical conductivity, which yields additional information about the recovery dynamics of the superconducting state. Fig. 4 shows the change in the conductivity $\Delta\sigma = \sigma(\Delta t) - \sigma_0$, where σ_0 is the equilibrium conductivity (without a pump pulse). $\Delta\sigma$ initially rises rapidly, as nonequilibrium quasiparticles are created and the superconducting state is depleted. After pumping, it decays. The overall timescale of this decay is given by the electron-phonon coupling, and thus faster for the breathing mode which is more strongly coupled to the electronic states. The decay is biexponential,

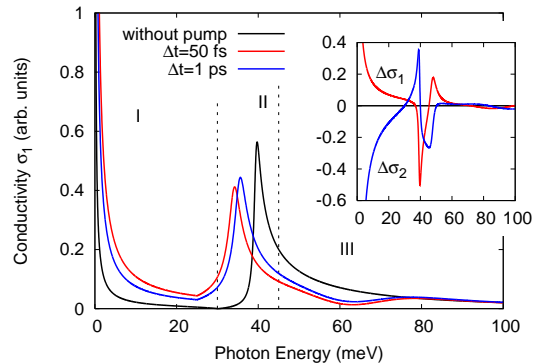


FIG. 2: Conductivity spectra for buckling (upper panel) and breathing (lower panel) modes. The equilibrium spectrum (without pump pulse) is shown along with spectra for different delay times. The inset shows the change in the real (red) and imaginary (blue) part of the conductivity, for $\Delta t = 0.5$ ps. It is $\Delta\sigma = \sigma(\Delta t) - \sigma_0$ with the equilibrium conductivity σ_0 .

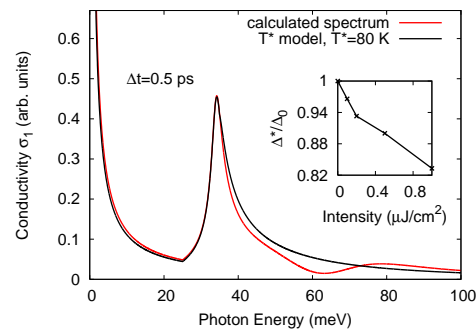


FIG. 3: Comparison of a calculated spectrum ($\Delta t = 0$) with a spectrum calculated using the effective T^* model. T^* is chosen in order to fit the calculated position of the pair-breaking peak. The inset shows the dependence of the T^* model gap Δ^* on the pump intensity.

with the two timescales corresponding to quasiparticle-phonon scattering and Cooper-pair recombination.

Derivation of rate equation approaches – Our microscopic approach can be used to derive a system of rate equations, which has been introduced by Kabanov *et al.* [1] to describe the combined dynamics of the excited quasiparticles and high-frequency phonons. The system is given by

$$\begin{aligned} \dot{n} &= I_0 + \eta N - R n^2 \\ \dot{N} &= J_0 - \eta \frac{N}{2} + R \frac{n^2}{2} - \gamma(N - N_T). \end{aligned} \quad (7)$$

n , N are the numbers of excited quasiparticles and phonons, respectively, η , R are rates denoting pair-breaking and Cooper-pair recombination, and I_0 , J_0 are the initial changes in n and N . N_T is the equilibrium phonon number, and γ describes phonon decay.

So far, we have only considered phonons within the bath approximation, where they remain in equilibrium.

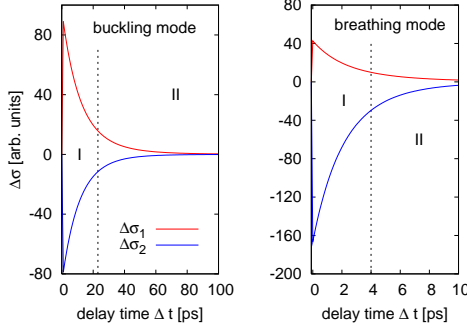


FIG. 4: Time evolution of the change in real (σ_1) and imaginary (σ_2) part of the optical conductivity for the buckling (left) and breathing (right) phonon modes. After an initial rise due to the pump process, a two-component decay of the conductivity follows. In region I scattering and in region II recombination dominate, respectively.

Our approach can be easily generalized to include the nonequilibrium phonon distributions within the Markovian approximation. The only modification in Eq. (6) is in fact that the phonon distributions $n_{\mathbf{q}j}$ are then time-dependent. A Boltzmann-like equation can also be derived for them. With $n \equiv \sum_{\mathbf{k}} (\langle \alpha_{\mathbf{k}}^+ \alpha_{\mathbf{k}} \rangle + \langle \beta_{\mathbf{k}}^+ \beta_{\mathbf{k}} \rangle) = 2 \sum_{\mathbf{k}} \langle \alpha_{\mathbf{k}}^+ \alpha_{\mathbf{k}} \rangle$, we can derive an equation for n by summing Eq. (6) over all \mathbf{k} . As only phonon absorption processes, i.e. pair-breaking by phonons, are relevant for Eq. (7), only the first (initial values) and the last two terms in (6) need to be considered. The first term gives an initial rate $I_0 \equiv -\frac{ie}{m} \sum_{\mathbf{k}} [\mathbf{k} \cdot \mathbf{A}_{\mathbf{q}} M_{\mathbf{k}\mathbf{q}} (\langle \alpha_{\mathbf{k}} \beta_{\mathbf{q}} \rangle - \langle \alpha_{\mathbf{k}}^+ \beta_{\mathbf{q}}^+ \rangle)]$. Assuming constant recombination and phonon absorption rates, $\Gamma_{\mathbf{k}\mathbf{q}j}^{(3)} \rightarrow \Gamma_{\mathbf{k}\mathbf{q}j}^{(3)}$ and $\Gamma_{\mathbf{k}\mathbf{q}j}^{(2)} \equiv n_{\mathbf{q}j} \tilde{\Gamma}_{\mathbf{k}\mathbf{q}j}^{(2)} \rightarrow n_{\mathbf{q}} \tilde{\Gamma}^{(2)}$, one directly gets the form of Eq. (7). A similar calculation with the phonon distribution equation yields the second rate equation. Thus, our microscopic approach includes the rate equations approach in the limit of constant scattering rates $\Gamma^{(i)}$. We can then write the rates R and η as:

$$R = \Gamma^{(3)}, \quad \eta = \tilde{\Gamma}^{(2)} \sum_{\mathbf{k}} \langle \alpha_{\mathbf{k}}^+ \alpha_{\mathbf{k}} \rangle (1 - \langle \alpha_{\mathbf{k}}^+ \alpha_{\mathbf{k}} \rangle), \quad (8)$$

where the rates $\Gamma^{(i)}$ are \mathbf{k} , \mathbf{q} , j -averages of the original rates, i.e. $\Gamma^{(i)} = \sum_{\mathbf{k}\mathbf{q}j} \Gamma_{\mathbf{k}\mathbf{q}j}^{(i)}$.

Conclusions – We have utilized density-matrix theory to calculate the ultrafast dynamics of high- T_c superconductors. Our novel microscopic description of the optical excitation includes both the depletion of the superconducting condensate, as well as relaxation of the excited quasiparticles and Cooper-pair recombination due to electron-phonon scattering. Pump-probe spectra, showing nonequilibrium effects above $2\Delta_0$, have been calculated as well as the real-time dynamics, where we find a biexponential decay produced by quasiparticle-

phonon scattering and Cooper-pair recombination processes. The relaxation times calculated for the buckling modes are compatible with experimental results [4]. We have compared our results with spectra calculated within the T^* model, finding good agreement in the low-energy limit, but our inclusion of nonequilibrium effects yields deviations at higher energies. Furthermore, we derive the widely used rate equation approaches from our microscopic formalism. Our method thus provides insight into the condensate dynamics of d -wave superconductors and includes earlier theoretical attempts to describe it.

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